

特許協力条約

PCT

特許性に関する国際予備報告（特許協力条約第二章）

（法第12条、法施行規則第56条）

〔PCT36条及びPCT規則70〕

出願人又は代理人 の書類記号 P798-PCT	今後の手続きについては、様式PCT/IPEA/416を参照すること。	
国際出願番号 PCT/JP2004/009466	国際出願日 (日.月.年) 28.06.2004	優先日 (日.月.年) 10.07.2003
国際特許分類 (IPC) Int.Cl. ⁷ C08L61/06, C08K3/22, C08L61/34		
出願人 (氏名又は名称) 旭有機材工業株式会社		

- この報告書は、PCT35条に基づきこの国際予備審査機関で作成された国際予備審査報告である。
法施行規則第57条（PCT36条）の規定に従い送付する。
- この国際予備審査報告は、この表紙を含めて全部で 3 ページからなる。
- この報告には次の附属物件も添付されている。
 - ☒ 附属書類は全部で 7 ページである。
 - ☒ 補正されて、この報告の基礎とされた及び／又はこの国際予備審査機関が認めた訂正を含む明細書、請求の範囲及び／又は図面の用紙（PCT規則70.16及び実施細則第607号参照）
 - ☐ 第I欄4.及び補充欄に示したように、出願時における国際出願の開示の範囲を超えた補正を含むものとこの国際予備審査機関が認定した差替え用紙
 - ☐ 電子媒体は全部で _____（電子媒体の種類、数を示す）。
配列表に関する補充欄に示すように、コンピュータ読み取り可能な形式による配列表又は配列表に関連するテーブルを含む。（実施細則第802号参照）

4. この国際予備審査報告は、次の内容を含む。

- ☒ 第I欄 国際予備審査報告の基礎
- ☐ 第II欄 優先権
- ☐ 第III欄 新規性、進歩性又は産業上の利用可能性についての国際予備審査報告の不作成
- ☐ 第IV欄 発明の単一性の欠如
- ☒ 第V欄 PCT35条(2)に規定する新規性、進歩性又は産業上の利用可能性についての見解、それを裏付けるための文献及び説明
- ☐ 第VI欄 ある種の引用文献
- ☐ 第VII欄 国際出願の不備
- ☐ 第VIII欄 国際出願に対する意見

国際予備審査の請求書を受理した日 12.04.2005	国際予備審査報告を作成した日 29.06.2005	
名称及びあて先 日本国特許庁 (IPEA/JP) 郵便番号100-8915 東京都千代田区霞が関三丁目4番3号	特許庁審査官 (権限のある職員) 吉宗 亜弓	4 J 3130
電話番号 03-3581-1101 内線 3457		

様式PCT/IPEA/409 (表紙) (2004年1月)

第I欄 報告の基礎

1. この国際予備審査報告は、下記に示す場合を除くほか、国際出願の言語を基礎とした。

☐ この報告は、_____ 語による翻訳文を基礎とした。

それは、次の目的で提出された翻訳文の言語である。

- ☐ PCT規則12.3及び23.1(b)にいう国際調査
☐ PCT規則12.4にいう国際公開
☐ PCT規則55.2又は55.3にいう国際予備審査

2. この報告は下記の出願書類を基礎とした。(法第6条(PCT14条)の規定に基づく命令に応答するために提出された差替え用紙は、この報告において「出願時」とし、この報告に添付していない。)

☐ 出願時の国際出願書類

☒ 明細書

第 1, 5-19 ページ、出願時に提出されたもの

第 2-4 ページ*, 12.04.2005 付けで国際予備審査機関が受理したもの

第 _____ ページ*, _____ 付けで国際予備審査機関が受理したもの

☒ 請求の範囲

第 9-13 項、出願時に提出されたもの

第 _____ 項*, PCT19条の規定に基づき補正されたもの

第 1-8 項*, 12.04.2005 付けで国際予備審査機関が受理したもの

第 _____ 項*, _____ 付けで国際予備審査機関が受理したもの

☐ 図面

第 _____ ページ/図、出願時に提出されたもの

第 _____ ページ/図*, _____ 付けで国際予備審査機関が受理したもの

第 _____ ページ/図*, _____ 付けで国際予備審査機関が受理したもの

☐ 配列表又は関連するテーブル

配列表に関する補充欄を参照すること。

3. ☐ 補正により、下記の書類が削除された。

☐ 明細書 第 _____ ページ

☐ 請求の範囲 第 _____ 項

☐ 図面 第 _____ ページ/図

☐ 配列表 (具体的に記載すること) _____

☐ 配列表に関連するテーブル (具体的に記載すること) _____

4. ☐ この報告は、補充欄に示したように、この報告に添付されかつ以下に示した補正が出願時における開示の範囲を超えてされたものと認められるので、その補正がされなかったものとして作成した。(PCT規則70.2(c))

☐ 明細書 第 _____ ページ

☐ 請求の範囲 第 _____ 項

☐ 図面 第 _____ ページ/図

☐ 配列表 (具体的に記載すること) _____

☐ 配列表に関連するテーブル (具体的に記載すること) _____

* 4. に該当する場合、その用紙に "superseded" と記入されることがある。

第V欄 新規性、進歩性又は産業上の利用可能性についての法第12条(PCT35条(2))に定める見解、それを裏付ける文献及び説明

1. 見解

新規性 (N)	請求の範囲	1-13	有
	請求の範囲		無
進歩性 (IS)	請求の範囲	1-13	有
	請求の範囲		無
産業上の利用可能性 (IA)	請求の範囲	1-13	有
	請求の範囲		無

2. 文献及び説明 (PCT規則 70.7)

- 文献1 : JP 2001-261976 A (大塚化学株式会社、河合石灰工業株式会社)
2001. 09. 26
文献2 : JP 5-279019 A (吉田工業株式会社) 1993. 10. 26
文献3 : JP 4-50105 A (日産化学工業株式会社) 1992. 02. 19

文献1, 2には、平均粒子径(短径)が100nm以下の板状ベーマイトを含有する樹脂組成物が記載されるものの、針状又は円筒状のベーマイトを含有することは、記載されていない。また、針状又は円筒状のベーマイトを樹脂に含有させることが、文献1, 2の記載から当業者にとって自明であるとも認められない。

また、文献3には、平均粒子径(短径)が100nm以下の針状ベーマイトを樹脂に含有し得ることが記載されるものの、該樹脂としてフェノール樹脂が記載されていない。一方、本願発明は、フェノール樹脂に平均粒子径(短径)が100nm以下で、針状又は円筒状であるという特定のベーマイトを含有させることによって、他のベーマイトを含有させた場合と比較して、機械的強度が向上するという顕著な効果を発揮する。

したがって、請求の範囲1-13に係る発明は、新規性及び進歩性を有する。

- 1 -

AMENDMENT
(Amendment Under PCT Article 34)

To: Examiner of the Patent Office: Junko Nakagawa

1. Identification of the International Application:
PCT/JP2004/009466

2. Applicant:
Name: ASahi ORGANIC CHEMICALS INDUSTRY CO., LTD.
Address: 5955, Nakanosecho 2-chome, Nobeoka-shi,
Miyazaki 882-8688 Japan
Country of nationality: JAPAN
Country of residence: JAPAN

3. Agent:
Name of Patent Attorney: (9975) AOKI Atsushi
Address: A. AOKI, ISHIDA & ASSOCIATES
Toranomon 37 Mori Bldg., 5-1, Toranomon
3-chome, Minato-ku, Tokyo 105-8423 Japan
Telephone 03-5470-1900

4. Items to be Amended:
Claims and Description

5. CONTENT OF AMENDMENT:

5-1. Specification:

(1) We amend the description, page 2, lines 10 and 11, "500nm to 1500nm" to "500nm to 15000nm".

(2) We amend the description, page 3, lines 23, 30 and 36, and page 4, line 13 "boehmite" to "acicular or cylindrical boehmite".

(3) We amend the description, page 4, line 35 to page 5, line 3, "The shape of the boehmite is not specifically limited and those having various shapes such as spherical, flat, acicular, cylindrical and amorphous are used. In view of availability and an increase in

mechanical strength, acicular or cylindrical boehmite is preferable." to "In view of availability and an increase in mechanical strength, the shape of boehmite is acicular or cylindrical.".

5-2. Claims:

(1) We amend claims 1 to 8, line 2 of each claim, "boehmite" to "acicular or cylindrical boehmite".

(2) We amend claim 6, line 6 and claim 8, line 7, "a filler" to "a filler". (No change in the English term. Only an amendment of Japanese character, "KANJI")

6. LIST OF ATTACHED DOCUMENTS

New pages of specification (page 2, page 3, page 3/1, page 4, page 5), one copy each.

New pages of claims (page 24, page 24/1, page 25), one copy each.

Fibrous fillers including glass fibers, which have conventionally been added so as to improve mechanical properties and heat resistance of a resin composition, had a problem such as difference in properties, particularly linear expansion coefficient, between molded article obtained in a flow direction during molding and a molded article obtained in a direction perpendicular to the flow direction (anisotropy). It is known to use, as the filler for solving the problem, boehmite having an external size of 0.5 to 15 μm (500 nm to 15000 nm) and an aspect ratio of 10 to 100 and to use a phenol resin as the resin (Japanese Unexamined Patent Publication (Kokai) No. 2001-261976). However, thermal conductivity, mechanical strength, kneading workability and moldability of these compositions remain to be improved.

Also there arose a problem that fluidity of the material deteriorates to thereby impair kneading workability and moldability, and thus there has been made a trial of securing fluidity of the material by using a benzooxazine resin, which has low melt viscosity before curing and is excellent in fluidity, as compared with a conventional phenol resin (Japanese Unexamined Patent Publication (Kokai) No. Hei 11-071498 and Japanese Unexamined Patent Publication (Kokai) No. 2001-064480). However, when the benzooxazine resin is used, the resulting resin composition has insufficient mechanical strength and a resin composition having better performances have been required.

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DISCLOSURE OF THE INVENTION

An object of the present invention is to provide a phenol resin composition which is excellent in thermal conductivity and mechanical strength.

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Another object of the present invention is to provide a (thermosetting) resin composition which is excellent in thermal conductivity and mechanical

strength, and also contains a phenol resin and a benzooxazine resin which are excellent in kneading workability and moldability.

5 The present inventors have intensively studied to achieve the objects described above and found that a phenol resin composition having excellent thermal conductivity and mechanical strength can be obtained by mixing the phenol resin with boehmite having a specific particle diameter. Thus the present invention has been
10 completed based on this finding.

Furthermore, the present inventors have intensively studied about the phenol resin composition so as to improve characteristics of the resin composition and found that a resin composition, which is excellent in
15 thermal conductivity and mechanical strength, and also contains a phenol resin and a benzooxazine resin which are excellent in kneading workability and moldability, can be obtained by mixing a resin mixture of a phenol resin and a benzooxazine resin with boehmite having a
20 specific particle diameter. Thus, an improved invention of the present invention has been completed.

That is, the phenol resin composition of the present invention comprises a phenol resin and acicular or cylindrical boehmite having an average particle diameter
25 (minor diameter) of 100 nm or less.

The resin composition containing a phenol resin and a benzooxazine resin of the present invention comprises a phenol resin and a benzooxazine resin in a weight ratio within a range from 95/5 to 25/75, and further comprises
30 acicular or cylindrical boehmite having an average particle diameter (minor diameter) of 100 nm or less.

EFFECT OF THE INVENTION

35 The phenol resin composition of the present invention has mechanical strength and thermal conductivity improved by mixing with acicular or cylindrical boehmite having an average particle diameter

(minor diameter) of 100 nm or

less, as compared with a conventional phenol resin composition, and is therefore preferably used for mechanical components, laminates and sheet materials, including molding materials for electrical and electronic components such as semiconductor sealing materials as well as molding materials for automobile components.

The resin composition containing a phenol resin and a benzooxazine resin of the present invention, which is excellent in mechanical strength and thermal conductivity and is also excellent in workability and moldability, is obtained by using a phenol resin and a benzooxazine resin in combination as a thermosetting resin and further adding acicular or cylindrical boehmite having an average particle diameter (minor diameter) of 100 nm or less, and is therefore preferably used for mechanical components, laminates and sheet materials, including molding materials for electrical and electronic components such as semiconductor sealing materials as well as molding materials for automobile components.

In the present invention, a novolak type phenol resin and a resol type phenol resin are used as the phenol resin and these resins are used alone or in combination. Among these resins, a novolak type phenol resin is preferably used. In this case, as a curing agent, hexamethylenetetramine is used in an amount of about 5 to 40 parts by weight based on 100 parts by weight of the novolak resin.

The boehmite used in the present invention is an inorganic compound represented by the general formula: $AlO(OH)$ which contains at least 90% or more aluminum hydroxide oxide. In the present invention, fine boehmite having an average particle diameter (minor diameter) of 100 nm or less is used. The average particle diameter is preferably from 1 to 100 nm, more preferably from 5 to 50 nm, and most preferably from 10 to 20 nm. The shape of the boehmite is not specifically limited and those having various shapes such as spherical, flat, acicular,

cylindrical and amorphous are used. In view of availability and an increase in mechanical strength, acicular or cylindrical boehmite is preferable. Furthermore, an aspect ratio (= average particle diameter of major diameter/average particle diameter of minor diameter) is preferably from 1 to 100, and more preferably from 5 to 50. In the present invention, boehmite having a size of 100 nanometers or less is referred to as "nanoalumina".

The amount of the nanoalumina in the present invention is appropriately decided according to the required physical properties and applications of the phenol resin composition, but is preferably from 1 to 150 parts by weight, and more preferably from 5 to 100 parts by weight, based on 100 parts by weight of the phenol resin. It is not preferable that the amount is less than 1 part by weight because performances such as mechanical strength and thermal conductivity are not sufficiently exhibited. It is not preferable that the amount is more than 150 parts by weight because fluidity deteriorates and thus it becomes difficult to perform kneading or molding.

In the present invention, when the composition further contains a benzooxazine resin, the amount of the nanoalumina is appropriately decided according to required physical properties and applications of the resin composition containing a phenol resin and a benzooxazine resin, but is preferably from 1 to 150 parts by weight, and more preferably from 5 to 100 parts by weight, based on 100 parts by weight the total amount of the phenol resin and the benzooxazine resin. It is not preferable that the amount is less than 1 part by weight because performances such as mechanical strength and thermal conductivity are not sufficiently exhibited. It is not preferable that the amount is more than 150 parts by weight because fluidity deteriorates and thus it becomes difficult to perform kneading or molding.

CLAIMS

1. (Amended) A phenol resin composition comprising a phenol resin and acicular or cylindrical boehmite having an average particle diameter (minor
5 diameter) of 100 nm or less.

2. (Amended) A phenol resin composition comprising a phenol resin and acicular or cylindrical boehmite having an average particle diameter (minor
10 diameter) of 100 nm or less, wherein the boehmite has an aspect ratio of 1 to 100.

3. (Amended) A phenol resin composition comprising a phenol resin, acicular or cylindrical boehmite having an average particle diameter (minor
15 diameter) of 100 nm or less and an alumina-based compound as a filler.

4. (Amended) A phenol resin composition comprising a phenol resin, acicular or cylindrical boehmite having an average particle diameter (minor
20 diameter) of 100 nm or less and an alumina-based compound as a filler, wherein the boehmite has an aspect ratio of 1 to 100.

5. (Amended) A phenol resin composition comprising a phenol resin and acicular or cylindrical boehmite having an average particle diameter (minor
25 diameter) of 100 nm or less, an amount of the boehmite being from 1 to 150 parts based on 100 parts by weight of the phenol resin.

6. (Amended) A phenol resin composition comprising a phenol resin and acicular or cylindrical
30 boehmite having an average particle diameter (minor diameter) of 100 nm or less, an amount of the boehmite being from 1 to 150 parts based on 100 parts by weight of the phenol resin, the phenol resin composition further comprising an alumina-based compound as a filler.

7. (Amended) A phenol resin composition comprising a phenol resin and acicular or cylindrical
35 boehmite having an average particle diameter (minor

diameter) of 100 nm or less and an aspect ratio of 1 to 100, an amount of the boehmite being from 1 to 150 parts based on 100 parts by weight of the phenol resin.

- 5 8. (Amended) A phenol resin composition comprising a phenol resin and acicular or cylindrical boehmite having an average particle diameter (minor diameter) of 100 nm or less and an aspect ratio of 1 to 100, an amount of the boehmite being from 1 to 150

parts based on 100 parts by weight of the phenol resin, the phenol resin composition further comprising an alumina-based compound as a filler.

5 9. The phenol resin composition according to any one of claims 1 to 8, which has thermosetting properties.

10 10. The phenol resin composition according to any one of claims 1 to 4, further comprising a benzoxazine resin in a weight ratio of the phenol resin to the benzoxazine resin within a range from 95/5 to 25/75.

15 11. The phenol resin composition according to any one of claims 5 to 8, further comprising a benzoxazine resin in a weight ratio of the phenol resin to the benzoxazine resin within a range from 95/5 to 25/75 (provided that the content of the boehmite is within a range from 1 to 150 parts by weight based on 100 parts by weight of the total amount of the phenol resin and the benzoxazine resin).

20 12. The phenol resin composition according to claim 10, which has thermosetting properties.

25 13. The phenol resin composition according to claim 11, which has thermosetting properties.